

# Direction dependency of extraordinary refraction index in uniaxial nematic liquid crystals

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## Abstract

The article presents a straightforward experiment which directly and illustratively demonstrates the double refraction. For this purpose two liquid crystalline cells were designed, which enable qualitative and quantitative measurements of the extraordinary refractive index direction dependency in an uniaxial nematic liquid crystal.

## I. TEACHING THE CONCEPT OF ANISOTROPY

Anisotropy of physical properties is a crucial property of materials which are nowadays extremely important in science and technology. An example of such materials are liquid crystals which enable production of several devices that are used in the everyday life (flat-screen TVs, notebooks, ipads, calculators, etc.). Even though students are in touch with such devices on a daily basis, they are not aware of the anisotropy as the key property of materials that enable the hi-tech products they use so eagerly. This ignorance might be due to the fact that the concept of anisotropy and in particular its consequences are rather difficult to comprehend.<sup>1</sup> It is thus important to introduce the concept of anisotropy as soon as possible, taking into account students' knowledge of physics and mathematics.<sup>2-5</sup>

In this paper we focus on the birefringence as one of the most widely used property of anisotropic materials. In birefringent material the velocity of light propagating through the material depends on the direction of propagation and polarization of light. Two distinct waves can propagate in a given direction. Both waves are linearly polarized, their polarizations are mutually perpendicular. They propagate with different phase velocities, which means that the refraction indices for the two waves are different. In general there are two directions of propagation in a given material along which the index of refraction is independent of polarization. These two directions are called optical axes and the material is said to be biaxial. When optical axes coincide, the material is uniaxial. In optically uniaxial materials one index of refraction is independent of the direction (the ordinary index of refraction) and the other one depends on the angle between the optical axis and the direction of light propagation (the extraordinary index of refraction). In optically biaxial materials both indices are in general angular dependent.

Most easily observed and striking optical property of transparent anisotropic materials is the double refraction (birefringence): the light incident on the interface between the isotropic and anisotropic material refracts into the anisotropic material such that there are in general two rays of light propagating through the material in different direction; both rays are polarized (even if the incident light is unpolarized), their polarizations are mutually perpendicular.

The double refraction is usually demonstrated by observing the doubling of a text observed through the calcite.<sup>6</sup> When a polarizer is placed behind the calcite (or in front of it),

one of the figures disappears if the polarizer's transmission direction coincides with one of the polarizations of the transmitted light. Although the explanation of the phenomenon is rather straightforward for a trained physicist, it is usually not so easily comprehended by students. A more straightforward experiment demonstrates the splitting of the non-polarized incident light beam into two perpendicularly polarized beams. One needs a large birefringent crystal, which is thick enough so that two transmitted beams can be observed as two light spots on a distant screen. Changing the direction of the incident light by rotating the crystal, the direction dependency of the extraordinary refraction index in uniaxial crystals as well as the direction dependency of both indices in biaxial crystals can be observed. Unfortunately, the accuracy of the quantitative measurements of both indices is poor, as any, even slight non-parallelism of the crystal surfaces results in significant changes of the transmitted light direction. Since the most easily accessible crystals are biaxial (e.g. quartz), this additionally complicates the comparison of the experimental results and the theoretical considerations.

Liquid crystals are optically anisotropic materials which are easy and cheap to obtain, even more, they can be synthesized in a school lab not only at the university level but also at the high school level. At the high school level the anisotropic properties can already be introduced through a set of carefully designed experiments. The concept can be efficiently reinforced at the university level, with an interdisciplinary teaching module consisting of lectures and lab work.<sup>3,7</sup> The module is appropriate for both social and natural science students. While such a teaching module proved to be very efficient in gaining the conceptual understanding of optical anisotropy, the Physics students require also experiments that would enable quantitative measurements. Liquid crystals offer a possibility to measure angular dependency of the extraordinary refractive index in uniaxial materials. In this paper we present an experiment which, besides the qualitative demonstration, enables quantitative measurements of the extraordinary refractive index for the light propagating at angles ranging from 0 to 90° with respect to the optical axis.

## II. LIQUID CRYSTALS AND THE DOUBLE REFRACTION

Nematic liquid crystals are composed of elongated molecules with orientationally ordered long molecular axes. The average direction of the long molecular axes is called the director. Due to the rapid molecular rotations around the long molecular axis, the system is opti-

cally uniaxial with the optical axis along the director. In bulky samples clusters of oriented molecules are formed and the director varies in space. In nematic liquid crystals the orientational correlation length, over which one can expect the same orientation of molecules, extends to  $500\text{ }\mu\text{m}$ , therefore the well-ordered samples have to be thinner than that.<sup>8</sup> Within the range of optical frequencies elongated molecules have larger polarizability along the long molecular axis than perpendicular to it, thus the speed of light is different for the two directions of polarizations. The light being polarized perpendicularly to the director is faster than the light having a component of polarization parallel to the director.

Propagation of light in anisotropic materials is described by the wave equation in which the anisotropic polarizability is described by the dielectric tensor ( $\underline{\varepsilon}$ ). By denoting the wave vector in the anisotropic material by  $\vec{k}$ , the magnitude of the wave vector in vacuum by  $k_0$  and the electric field (which also gives the direction of polarization) by  $\vec{E}$ , the wave equation can be written as<sup>9</sup>

$$\vec{k} \times (\vec{k} \times \vec{E}) + k_0^2 \underline{\varepsilon} \vec{E} = 0 \quad . \quad (1)$$

For ordinary nonabsorbing materials the dielectric tensor is symmetric, so there always exist a coordinate system with a set of axes, called the principal axes, in which the dielectric tensor is diagonal. In optically uniaxial materials with the optical axis in the  $z$ -direction, the  $x$ - and  $y$ -components of the dielectric tensor are equal:

$$\underline{\varepsilon} = \begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix} \quad ,$$

where  $n_o$  is called the ordinary and  $n_e$  the extreme extraordinary index of refraction. For each direction of the wave propagation there exist two solutions for the magnitude of the wave vector  $k$ . Assuming  $\vec{k} = (k_x, 0, k_z)$  and solving the wave equation (1) one finds:

$$k_x^2 + k_z^2 = k_0^2 n_o^2 \quad (2)$$

and

$$\frac{k_x^2}{n_e^2} + \frac{k_z^2}{n_o^2} = k_0^2 \quad . \quad (3)$$

The first solution (Eq. (2)) presents the ordinary wave, as for each direction of  $\vec{k}$  the index of refraction is the same ( $n_o$ ). The second one (Eq. (3)) presents the extraordinary wave. The magnitude of the wave vector and thus the index of refraction experienced by light, depend on the direction of  $\vec{k}$ ; the allowed values of  $k$  lie on the ellipse (Fig. 1). From the wave equation it also follows that the two allowed waves are linearly polarized, the extraordinary wave in the plane defined by the optical axis and  $\vec{k}$  and the ordinary wave in the direction perpendicular to this plane.

Expressing the magnitude of the wave vector of the extraordinary wave as  $k = k_0 n_e(\theta)$ , where  $n_e(\theta)$  is the extraordinary index of refraction for the light propagating at an angle  $\theta$  with respect to the optical axis, the angular dependence of the extraordinary index of refraction can be derived from Eq. (3):

$$n_e^2(\theta) = \frac{n_e^2 n_o^2}{n_o^2 \sin^2 \theta + n_e^2 \cos^2 \theta} . \quad (4)$$

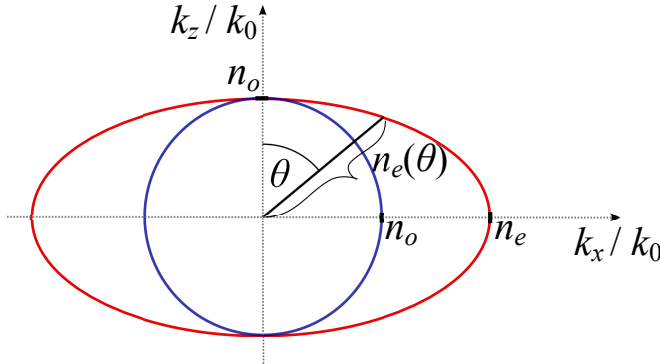


FIG. 1. Wave-vector surfaces for a point-like light source within an uniaxial anisotropic material. The wave-vector surfaces are obtained by the rotation of the circle and the ellipse around the optical axis which is along the  $z$ -axis.  $\theta$  is the angle between  $\vec{k}$  and the optical axis,  $n_o$  is the ordinary and  $n_e$  is the extreme value of extraordinary refractive index and  $n_e(\theta)$  the value of the extraordinary refractive index at an angle  $\theta$  with respect to the optical axis.

When the unpolarized light beam is incident on the anisotropic uniaxial transparent medium, it refracts into two light beams; since the refractive indices for the two beams are different so are the refractive angles. The ordinary and the extraordinary beam are linearly polarized, perpendicularly to each other. The direction of the extraordinary beam in the birefringent material depends on the incident angle and the orientation of the optical axis

with respect to the surface normal (Fig. 2).

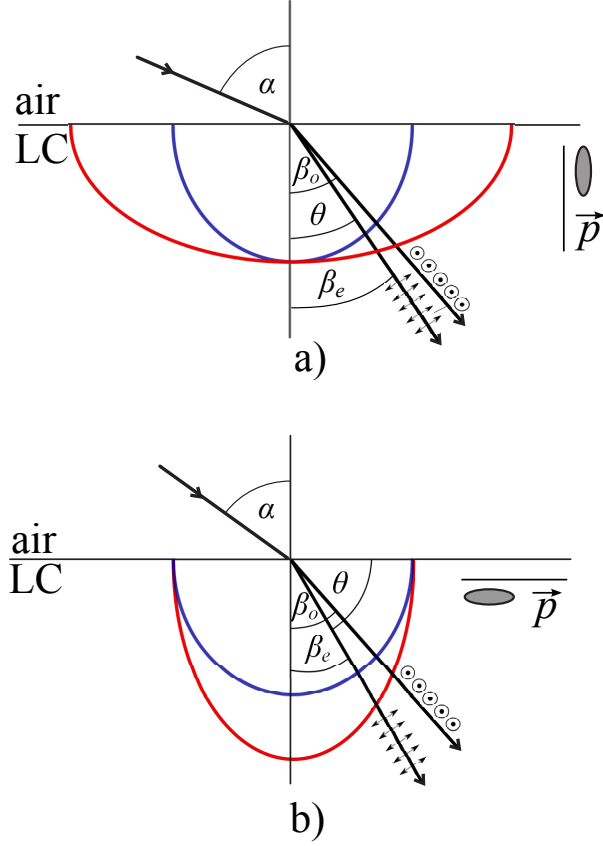


FIG. 2. Polarizability of an anisotropic material depends on the direction of the light propagation. Optical axis  $\vec{p}$  is a) perpendicular to the surface and b) along the surface in the incidence plane (the plane defined by the wave vector and the optical axis). The incident angle is  $\alpha$ ,  $\beta_o$  and  $\beta_e$  are the refraction angles of the ordinary and extraordinary wave, respectively, and  $\theta$  is the angle between the wave vector and the optical axis. Grey ellipsis marks orientation of elongated liquid crystalline molecules.

### III. EXPERIMENT

To provide the refraction situations shown in Fig. 2 two wedge cells were designed with different surface treatment in order to achieve different orientations of the director in the cell and thus different orientation of optical axis with respect to the surface (Fig. 3). The cells were approximately 1 cm long ( $h$ ) and half a centimeter wide (Fig. 4a). Usual laser beams are too wide to allow studies of double refraction in cells having parallel surfaces.

In thin samples, which guarantee the homogeneity of the orientation of long molecular axis (the director gives the direction of the optical axis), the spatial separation of the ordinary and extraordinary beam can be obtained by the prismatic effect.<sup>1,10</sup> To prepare a wedge cell a foil with thickness  $d = 360 \mu\text{m}$  was inserted and glued between two pieces of microscope glasses in one of the narrower sides, while the other narrow side of the cell was glued together directly, thus forming a wedge. By rubbing the surfaces the planar cell in which molecules are aligned in the surface plane along the long side of the surface (Fig. 3b) was designed. The elongated molecules align with their long axes along the scratches and the director is parallel to the rubbing direction. Therefore the optical axis also coincides with the rubbing direction. To align the molecules perpendicular to the surface (homeotropic cell, Fig. 3a) a polymer coating was applied to the glass. Professional cells usually use ITO treatment but for simple experiments the satisfactory effect is provided by simple dipping of the glass into the detergent or lecithin solution. The capillary effect is used to fill the cells by the liquid crystal E18<sup>11</sup> heated above the transition temperature from the nematic to the isotropic phase. For E18 the values of the ordinary and extraordinary indices are  $n_o = 1.52$  and  $n_e = 1.75$ .<sup>11</sup>

The experimental setup is shown in Fig. 4a. The wedge cell is fixed into the holder and it is put on the rotatable table with the longer side parallel to the table surface. A He-Ne laser is used as a source of the unpolarized light beam. The direction of the incident light is always in the incident plane perpendicular to the wedge. When a laser beam of unpolarized light is incident on the wedge cell, two bright spots are observed on a distant screen due to the birefringence of the liquid crystal in the wedge cell. The ordinary and the extraordinary refraction index are determined from the measurements of the relative position of the spots to the position of the direct beam spot, when light does not pass through the cell (Fig. 4b). The position of spots changes by changing the incident angle of light by rotating the table, which enables measurements of the angular dependence of both indices.

Fig. 5 shows the geometry of the light propagation through the wedge cell. The angle  $\alpha$  is the controlled incident angle and  $\beta_o$  and  $\beta_e$  are the refraction angles of the ordinary and the extraordinary polarized light, respectively. Both refraction angles are obtained from the direction given by  $\gamma$  of the refracted light (see Fig. 5), which can be calculated from the positions of the light dots on the screen ( $x_o$  and  $x_e$ ), the distance  $l$  between the cell and the screen and the wedge angle  $\delta$  (Fig. 4b) which is approximately  $d/h$  (Fig. 3). Since the  $l$  is

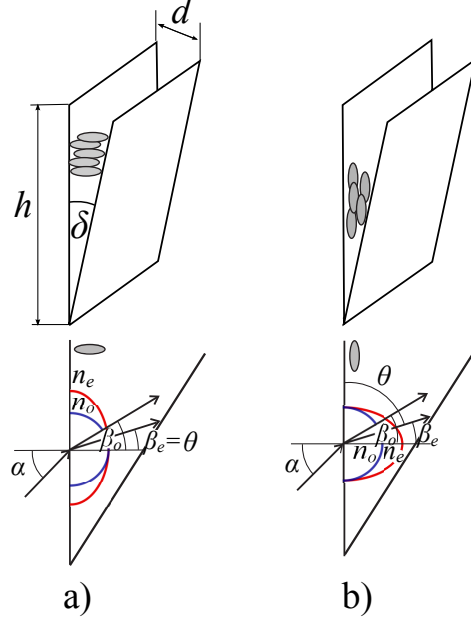


FIG. 3. a) In the homeotropic cell molecular long axes are oriented perpendicularly to the cell glass. The extraordinary index increases with the increasing incident angle  $\alpha$  (below). The wedge of the cell is defined by angle  $\delta$  which is a characteristic of a certain cell calculated from the length of the cell  $h$  and the thickness of the foil  $d$ . b) In the planar cell molecular long axes are oriented parallel to the longer side of the cell. The extraordinary index decreases from its maximum value when the incident angle increases. The angles are defined in Fig. 2.

large in comparison to the dimensions of the cell one can measure it as a distance between the spot position on the wall and the position of the beam in the cell.

Using the identity

$$\frac{\sin \alpha}{\sin \beta} = \frac{\sin(\alpha \pm \gamma \pm \delta)}{\sin(\beta \pm \delta)} = n \quad , \quad (5)$$

where  $n$  is the refractive index of either ordinary or the extraordinary wave. In Eq. (5) the upper sign in  $\pm$  stands for the beam direction given in Fig. 5 by the solid line and the lower sign for the beam denoted by the dashed line. Eq. (5) is an approximation since the direction of the optical axis in the cell slightly varies due to the wedge. However, since  $\delta$  is small, one can assume that the extraordinary index of refraction at angles  $\beta$  and  $\beta \pm \delta$  are the same. This is confirmed by the measurements where  $x_e$  measured at incidence angles  $\pm\alpha$  are the same within the experimental error and the width of the spot on the screen.

Assuming that  $\delta \ll 1$  rad, the refracted beam direction  $\beta$  in the anisotropic liquid crystal



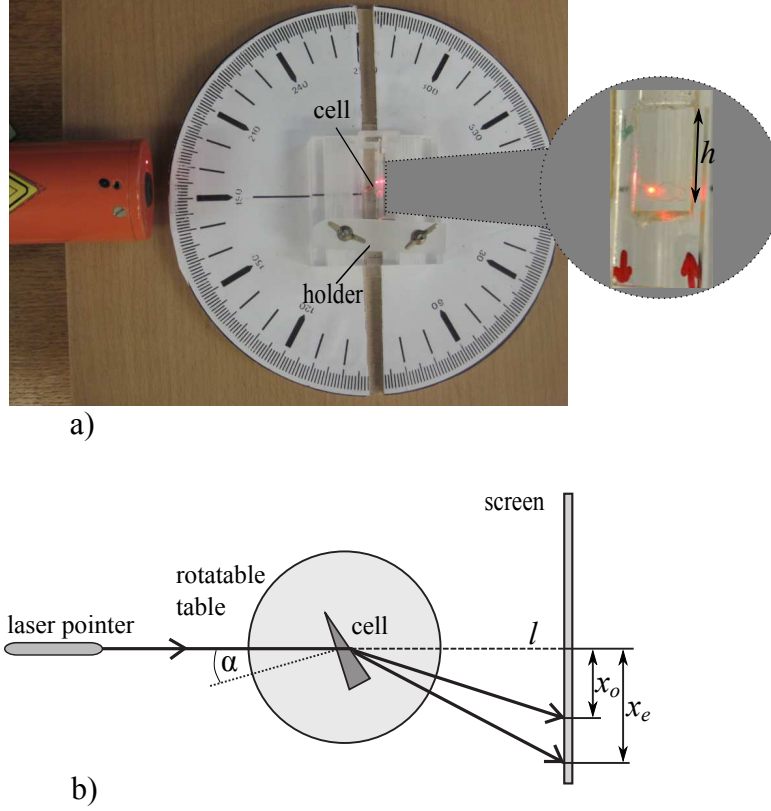


FIG. 4. a) (*Color online*) Experimental setup with a zoomed liquid crystalline cell. b) Schematic presentation.  $l$  is the distance between the cell and the screen.  $x_o$  and  $x_e$  define the position of the ordinary and extraordinary beam, respectively. For example, if one takes a planar cell with a wedge angle  $\delta = 3.2^\circ$ , then at the incident angle  $\alpha = 10^\circ$  and  $l = 5.07$  m the ordinary and the extraordinary beams hit the screen at  $x_o = 15.5$  cm and  $x_e = 22.0$  cm, respectively.

in the wedge is found:

$$\tan \beta = \frac{\delta}{\delta + \gamma} \tan \alpha \quad , \quad (6)$$

With  $\beta$  obtained from Eq. (6) the value of the refraction index is found from Eq. (5). Eqs. (5) and (6) are general expressions which can be used to obtain the refraction index when light passes through the thin wedge sample of any, not necessary birefringent, material and they enable the evaluation of both, the ordinary and the extraordinary refraction indices.

From Fig. 2 it is clearly seen that the polarizability depends on the direction of light propagation only if the light propagates in the plane defined by the optical axis and the surface normal. In the homeotropic cell (Fig. 3 a), the optical axis is perpendicular to the

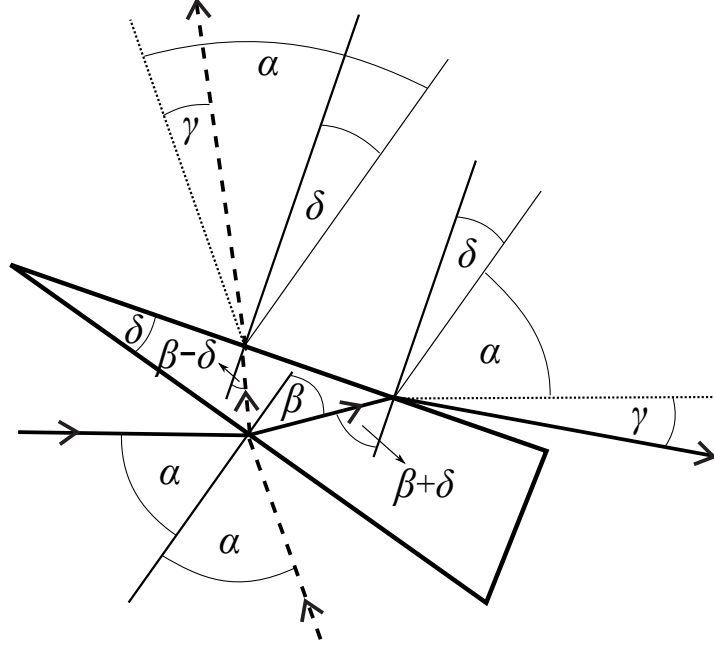


FIG. 5. The geometry of the experiment. The angles are defined in the text.

glass plate and the refraction angle  $\beta_e$  is equal to the angle  $\theta$  between the optical axis and  $\vec{k}$ , so:

$$\frac{\sin \alpha}{\sin \beta_e} = n_e(\theta) \quad , \quad (7)$$

where  $\beta_e$  is calculated from Eq. (6). When the direction of incident light is perpendicular to the cell, the electric field is perpendicular to the optical axis and the beam does not split. However, any departure from the perpendicular incidence gives a component of the electric field along the long molecular axis and consequently the value of the extraordinary index changes from the value of the ordinary index (Fig. 3a). From the measurements of the spot positions on the screen the values of the extraordinary refractive index at angles ranging from  $0^\circ$  to approximately  $40^\circ$  are calculated.

In the planar cell, the optical axis is parallel to the glass and the refraction angle is related to the angle between the optical axis and the wave vector direction as  $\beta_e = \frac{\pi}{2} - \theta$ . The extraordinary refractive index is therefore given by:

$$\frac{\sin \alpha}{\sin \beta_e} = \frac{\sin \alpha}{\sin (\frac{\pi}{2} - \theta)} = n_e(\theta) \quad . \quad (8)$$

From these measurements the values of the extraordinary refractive index at angles ranging

from approximately  $60^\circ$  to  $90^\circ$  are obtained. The planar cell can thus be used to study the direction dependency of the extraordinary index when the difference of both indices is close to its largest value.

There are few limitations in the experiment which have to be considered. Although the light with the ordinary and extraordinary polarization can propagate in any direction, experimentally we are limited with the refraction of the incident light, since the light source is outside the birefringent material. In the presented situation the ordinary beam direction was theoretically limited (for the light parallel to the cell surface) to  $41^\circ$  for  $n_o = 1.52$  and to  $35^\circ$  for  $n_e = 1.75$ . The cell size and the cell holder additionally limit the incident angle  $\alpha$ . In Fig. 6 we show the combined results of the extraordinary and ordinary index measurements in the planar and the homeotropic cell and compare them to the theoretical values which were calculated from Eq. (4) using the known values of both indices.

From Fig. 6 it is clearly seen that with such a simple experiment we were not able to measure the values of the extraordinary refraction index for directions around  $45^\circ$  with respect to the optical axis. To measure the extraordinary refraction index also for these directions one should use an old experimental trick - the phase matching by the help of an additional material.<sup>13</sup> In order to enter the liquid crystal under a general angle one should prevent the refraction between two materials which have a large difference in refraction indices. Therefore the light should pass the surface where optical indices of materials on both sides of the surface are similar.

To achieve such conditions the wedge cell is sandwiched between two glass (refraction index  $n_g = 1.50$ ) prisms with the apex angle  $45^\circ$  (Fig. 7). To prevent the existence of a tiny air interface between the prism and the wedge cell, the contact areas are covered by glycerol, which has the refraction index similar to the refraction indices of glass and liquid crystal.<sup>12</sup> If the incident angle on the prism surface is zero, the beam does not refract and the incident angle to the liquid crystal in the wedge is  $45^\circ$  (Fig. 8). As the refraction indices of the glass and liquid crystal are similar, the angle of refraction does not differ much from the incident angle and in the liquid crystal light propagates in the direction close to  $\theta = 45^\circ$ . The prism at the other side of the cell provides the opposite change of the light direction as the first one. Without the wedge cell the light beam should be straight. Therefore the positions of the two light spots again enable the measurement of both refraction indices simultaneously.

Let us follow the light beam through the sandwich and calculate the refraction indices

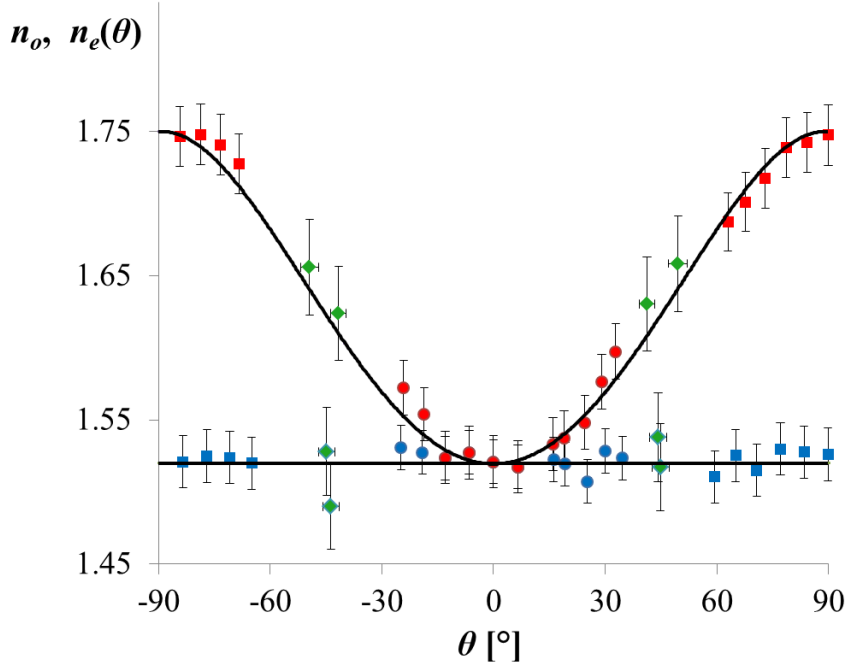


FIG. 6. (*Color online*) The refractive indices ( $n_o$  and  $n_e(\theta)$ ) as a function of the angle  $\theta$  between the optical axis and wave vector in nematic liquid crystal E18. Squares: values obtained from the measurements in the planar cell. Circles: values obtained from the measurements in the homeotropic cell. Red: extraordinary, Blue: ordinary index of refraction. Green diamonds: measurements of ordinary and extraordinary index of refraction in the sandwiched cell (see Fig. 8). The theoretical direction dependence for  $n_e = 1.75$  and  $n_o = 1.52$  is given for a comparison.

(Fig. 8). If the glycerol film is of uniform thickness it does not influence the analysis of the refraction indices. The first relevant interface is thus glass - liquid crystal. The incident angle is  $\alpha = 45^\circ$  and the beam refracts only slightly ( $\beta$  is close to  $45^\circ$ ), because the refraction indices of liquid crystal are close to 1.5. At the liquid crystal - glass interface the incident angle changes due to the wedge angle  $\delta$  to  $\beta + \delta$  as in Fig. 8 or to  $\beta - \delta$  if the sandwich is rotated for  $90^\circ$  clockwise. Therefore it can be written

$$\frac{\sin(\beta \pm \delta)}{\sin \beta'} = \frac{n_g}{n}, \quad (9)$$

where  $\beta'$  is the refractive angle defined in Fig. 8. The last refraction occurs at the glass - air interface. The incident angle equals to  $\beta' - \pi/4$  as in Fig. 8 or  $\beta' + \pi/4$  if the sandwich is rotated for 90 degrees clockwise as in Fig. 7. One can write the Snell's law as

$$\sin(\beta' \mp \pi/4) = \frac{\sin(\gamma \pm \delta)}{n_g} , \quad (10)$$

where  $\gamma \approx x/l$  is again the angle between the incident unpolarized light and the direction of light after passing the sandwich of prisms and the cell.

The angle  $\theta$  between the optical axis and the extraordinary/ordinary ray direction is equal to  $\beta$  in the homeotropic cell and  $\pi/2 - \theta$  in the planar cell (see Fig. 3).

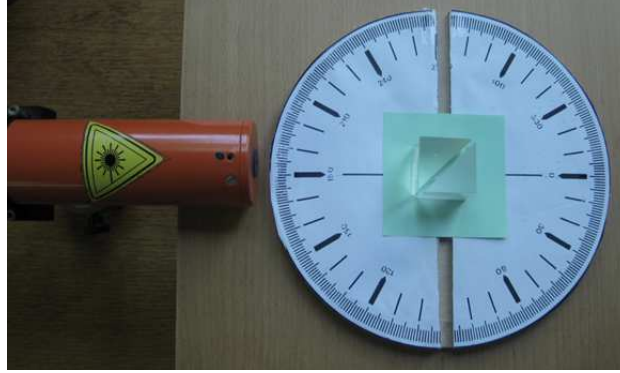


FIG. 7. (*Color online*) The sandwich of prisms and the cell.

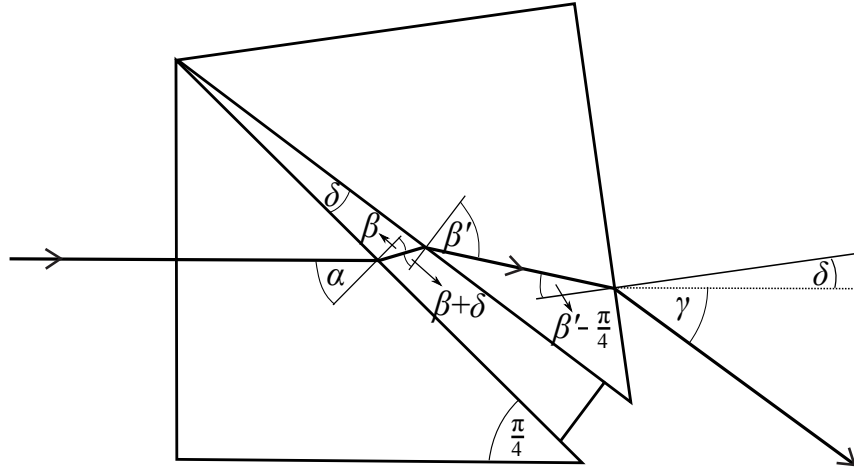


FIG. 8. The geometry of the upgraded experiment (sandwich). The angles are defined in the text.

As explained above, the sandwiched cells provide extra measurements of refractive indices at angles  $\theta$  close to  $45^\circ$ . Since we have two cells with different alignments of molecules and two different orientations of sandwich as in Figs. 7 and 8, the refraction indices in four different directions of light propagation in the cell with respect to the optical axis can be measured. These measurements are shown as green diamonds in Fig. 6. One can clearly see

that the accuracy of the refraction index measurement when the wedge cell is fixed between the prisms is much lower than without them. The reason is the glycerol, because it forms a slight wedge as well, which could not be precisely controlled. The wedge of the glycerol has in most cases the same orientation as the wedge of the liquid crystalline cell, which results in larger values of refractive indices. Less frequently the glycerol wedge is in the opposite direction which leads to lower values of the refractive index. The effect was verified in the absence of the liquid crystal. Nevertheless, the experiment nicely shows that values of the refractive indices at  $\theta$  between  $40^\circ$  and  $50^\circ$  are consistent with the calculated direction dependence of the extraordinary refraction index in the uniaxial liquid crystal.

#### IV. CONCLUSION

Students are confronted by a difficult concept of birefringence during the physics lessons at the university. We have shown that the concept can be fully demonstrated by using the nematic liquid crystalline wedge cells with different orientation of molecules. Most important, the experiment, whose setup consists of a laser, rotatable table, two different wedge cells, two prisms with the apex angle  $45^\circ$  and a drop of glycerol, enables quantitative measurements of the angular dependence of the extraordinary refractive index for the whole range of directions. The homeotropic cell enables the demonstration of the direction dependency of the extraordinary index, as well as its measurement, when its value is close to the value of the ordinary refraction index. The planar cell can be used for the quantitative measurement of the extraordinary index direction dependency close to its largest value. By additional glass prisms forming the sandwich of prisms and the wedge cell one can study the angular dependency of the extraordinary refractive indices in the range of directions that cannot be reached by a simple setup.

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- <sup>10</sup> D. K. Shenoy, "Measurements of Liquid Crystal Refractive Indices," *Am. J. Phys.* **62**, 858-59 (1994).
- <sup>11</sup> Nematic liquid crystals with appropriate properties are commercially available by Merck.
- <sup>12</sup> Refractive index of glycerol, <http://refractiveindex.info/>.
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